Plasticity of Ferromagnets near the Curie Point

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We propose an explanation of the anomalous growth of plasticity in ferromagnets near the Curie point. We demonstrate that this effect is caused by spin-dependent detachment of dislocations from obstacles under an influence of the internal magnetic field. Magnetization fluctuations grow in the vicinity of the Curie point, yield an increase of the detachment probability and, hence, an increase of the plasticity. We apply this model for a description of the temperature behaviour of the critical stress in nickel and of the microhardness of gadolinium. An external magnetic field suppresses the magnetization fluctuations and, hence may suppress the above singularities.

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I. INTRODUCTION

Various experimental data indicate a strong influence of the magnetic structure of ferromagnets on their plastic properties. Transition to a magnetically ordered state is accompanied by a strong change of plasticity (Zackay and Hazlett, 1953, Nabutovskaya, 1969,1971, Wolfenden, 1978, Bolling and Richman, 1969, Echigoya etal, 1973, Echigoya and Hayashi, 1979, Flor etal, 1980, Retat etal, 1985, Retat, 1987, Maksimova and Maiboroda, 1992, Gulayev and Svistunova, 1996). Measurements with small temperature intervals revealed an increase of plasticity in a close vicinity of the Curie point (Zackay and Hazlett, 1953, Nabu-

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tovskaya, 1969,1971, Wolfenden, 1978). This fact by itself is not so astonishing since many properties of ferromagnets change near the Curie point (Vonsovskii, 1974), however, the giant value of the observed effect is really surprising. According to Nabutovskaya (1969,1971) the microhardness of gadolinium changes near T_C by a factor of two, whereas its elastic constants vary by a percent or so. For example, the Young modulus of gadolinium decreases in the Curie point only by 1.6% (Spichkin etal, 1999).

Recently we considered a mechanism of the influence of a magnetic field on plasticity of some nonmagnetic crystals (Molotskii and Fleurov, 1997, Molotskii, 2000, and references therein). The magnetic field induces transitions (singlet to triplet) between different spin states of the radical pairs formed by dangling bonds of a dislocation cores and paramagnetic obstacles in the course of the pair formation. The state of the pair may be either bonding or antibonding, depending on its spin configuration. The magnetic field can influence the relative occupations of these states and lead to an increase of probability of the dislocation detachment from paramagnetic obstacles. The crystal plasticity, as a result, increases.

This approach appeared to be very successful and allowed for an explanation of a large number of plasticity related phenomena in nonmagnetic crystals. We propose here to apply the same approach when discussing plasticity of ferromagnets. In this case, the role of the external magnetic field can be played by the local magnetic induction created by the spontaneously magnetised surrounding atoms. Recently, we demonstrated that this internal magnetic field may be important for the Invar hardening, leading to an increase of the critical resolved shear stress of Invar alloys with a lowering temperature (Molotskii and Fleurov, 2001). We expect to observe a temperature variation of various plasticity related characteristics of the crystal connected with the variation of the spontaneous magnetization with the temperature. When approaching the Curie point the role of the magnetization fluctuations may become more pronounced. The typical time of relaxation of the magnetization fluctuations is usually much larger than the characteristic times of formation or rupture of a dislocation - obstacle bond. Therefore we should consider this process under the influence of slowly varying magnetic inductions due to fluctuating magnetization. We will show below that this may result in a strongly increasing plasticity in the vicinity of the Curie point.

It is worth mentioning here that the theory, we develop in this paper, will not take into account an interaction of dislocations with domain walls, which may serve as efficient pinning centres for dislocation (Seeger etal, 1964). This assumption is justified as long as the typical

domain size essentially exceeds the dislocation path length. According to the experiments of Zackay and Hazlett (1953), and of Nabutovskaya (1969, 1971) this path is limited by the average distance, $L_f \sim 1/\sqrt{\rho_f}$, between the forest dislocations. The density of the forest dislocations in those experiments was $\rho_f \sim 10^9 \text{cm}^{-2}$, hence the dislocation path length could not exceed $L_f \sim 3 \times 10^{-5}$cm, which was two to three orders of magnitude smaller than the typical domain sizes.

II.   INFLUENCE OF INTERNAL MAGNETIC INDUCTION ON PLASTICITY OF MAGNETS

Internal magnetic field of a ferromagnet due to spontaneous magnetization is rather large ($\sim$ 1T) and is capable of influencing plastic properties of crystals. Such an important characteristic of crystal plasticity as the dislocation path length increases with the magnetic induction B as (Molotskii, 2000)

$$L(B) = L_0(1 + \frac{B^2}{B_0^2}) \tag{1}$$

where B_0 is a constant characterizing the dislocation – obstacle bond. Usually its value lies in the range from 0.2 to 1T. In the case of nonmagnetic crystals B is the value of the magnetic induction created by an external source. In the case of a ferromagnet we do not need an external magnetic field since a strong enough internal magnetic induction is always present. This internal magnetic induction decreases when temperature approaches the Curie point T_C and starts strongly fluctuating. One may expect that in the critical region, at $T \to T_C$, these strong fluctuations of the internal magnetic induction will strongly influence plastic properties of the ferromagnet.

The magnetic field influences kinetics of the formation of the dislocation – obstacle bonds. The characteristic times of these processes are typically on the order of 10^{-7}s (Molotskii and Fleurov, 1997), which is shorter than the typical times of the large scale magnetization fluctuations determining the local internal magnetic induction B_{int} at each bond (see, e.g., Ma, 1976). This allows one to consider the induction B_{int} at each moment of time as created by a particular space distribution of the magnetization fluctuations in the ferromagnet.

For example, we estimate here the average dislocation free path length. According to

equation (1) it is determined by the mean square of the internal magnetic induction,

$$\langle B_{int}^2 \rangle = \overline{B}_{int}^2 + \langle \Delta B^2 \rangle. \tag{2}$$

Here $\overline{B}_{int}$ is average value of the internal magnetic induction, whereas $\Delta B = B_{int} - \overline{B}_{int}$.

Fluctuations of the local internal magnetic induction acting on a particular dislocation – obstacle bond can be described using the theory of the second order phase transitions (see, e.g., Landau and Lifshitz, 1980). The local internal magnetic indiction of a homogeneously magnetised crystal can be estimated with the help of the Lorentz formula

$$\mathbf{B} = \frac{4\pi}{3}\mathbf{M}. \tag{3}$$

The probability that the magnetization M deviates by ΔM from its average value $\overline{M}$ in the volume V around the bond is

$$w(\Delta M, V) = \frac{1}{2}\frac{V^{*1/2}}{(2\pi k_B T\chi)^{3/2}} \left\{ \begin{array}{ll} \exp\left\{-\dfrac{\Delta \mathbf{M}^2 V}{2k_B T_C \chi}\right\}, & V > V^* \\[2mm] 0, & V < V^* \end{array} \right. \tag{4}$$

where χ is the magnetic susceptibility, k_B is Boltzmann constant. The probability density function (4) is truncated at volumes smaller than certain characteristic volume V^* which will later serve as a fitting parameter. The general approach to the order parameter fluctuations, applied here, considers the ferromagnet as continuus medium. Hence, it holds at distances which are larger than typical interatomic distances. Therefore one may expect the typical scale of the excluded volume V^* not to exceed essentially several lattice spacings. Similar approach is implied by equation (3), which is obtained by considering a small empty volume surrounded by a magnetised medium.

The mean square fluctuations of the magnetization can be then calculated as

$$\langle \Delta \mathbf{M}^2 \rangle = \int_{V^*}^{\infty} d^3\Delta M (\Delta \mathbf{M})^2 w(\Delta M, V) = \frac{k_B T_C \chi}{V^*} \tag{5}$$

Now using equations (2) and (5) the mean square local magnetic field acting on the dislocation – obstacle bond can be obtained using the following equations

$$\langle \mathbf{B}_{int}{}^2 \rangle = \overline{B}_{int}{}^2 + \langle \Delta B^2 \rangle = \frac{16\pi^2}{9}\overline{\mathbf{M}}{}^2 + \frac{16\pi^2}{9}\frac{k_B T_C \chi}{V^*}. \tag{6}$$

An inhomogeneous spatial distribution of the magnetization, caused by fluctuations, results in a demagnetization field, which should have been accounted for in equation (3). The

demagnetization field is the strongest for the spherical fluctuations and is negligible for a fluctuation having the form of a plate. The exact account of the demagnetization is a very tedious task but finally leads to a numerical factor, smaller than one, in the second term in equation (6). This factor is absorbed in the volume V^*, which anyhow serves as a fitting parameter.

The temperature dependence of the mean square local magnetic field in the vicinity of the Curie point can be readily calculated. For this we may use the temperature dependence of the magnetic susceptibility which has the form

$$\chi(T) = \begin{cases} \dfrac{C}{2T_C} \left(\dfrac{T_C}{T_C - T}\right)^{\gamma}, & \text{at} \quad T < T_C \\[4mm] \dfrac{C}{T_C} \left(\dfrac{T_C}{T - T_C}\right)^{\gamma} & \text{at} \quad T > T_C. \end{cases} \tag{7}$$

Here γ is a critical index,

$$C = \frac{np^2\mu_B^2}{3k_B} \tag{8}$$

is the Curie constant, in which n is the particle density, p is the effective number of the Bohr magnetons μ_B per atom.

The first term in equation (6) contains the average spontaneous magnetization $\overline{M}(T)$ whose temperature dependence is

$$\overline{M}(T) = \begin{cases} M_0 \left(1 - \dfrac{T}{T_C}\right)^{\beta} & \text{at} \quad T < T_C, \\[4mm] 0 & \text{at} \quad T > T_C. \end{cases} \tag{9}$$

Here M_0 is the limiting value of the magnetization at low temperature, β is the corresponding critical index

III. TEMPERATURE DEPENDENCE OF THE CRITICAL STRESS: NICKEL AS A TEST CASE

An influence of the magnetization on plasticity was first discovered half a century ago by Zackay and Hazlett (1953), who studied the temperature dependence of the critical stress σ_c for nickel. They have found that the dependence of $\sigma_c(T)$ passes a minimum near the Curie point T_C. Samples with 99.95% nickel content were used. There was also a small amount

of iron (0.03%) and magnesium (0.02%) atoms. The paramagnetic iron atoms are efficient obstacles for dislocations in nickel, and they are most probably responsible for the observed sensitivity of the plasticity of nickel to the magnetic field.

The temperature dependence of the critical stress in a ferromagnet can be found, accounting for the part played by the internal magnetic fields. Dislocations can be bound to paramagnetic obstacles with the binding energy W_M. Then the critical stress is (Friedel, 1964)

$$\sigma_c(0) = \frac{|W_M|}{b^2 l(B)} \left[1 - \left(\frac{T}{T_0} \right)^{\frac{2}{3}} \right]^{\frac{3}{2}} \tag{10}$$

where b is the value of the dislocation Burgers vector, and l is the average distance between the obstacles. The temperature dependent factor in equation (10) accounts for the thermal activation processes (Haasen, 1983). The parameter T_0 is proportional to the dislocation - obstacle binding energy.

It was shown in our paper (Molotskii and Fleurov, 1997) that the average length of the dislocation free segment depends on the magnetic field similarly to (1),

$$l(B) = l_0 \left(1 + \frac{B^2}{B_0^2} \right). \tag{11}$$

Considering a ferromagnet we should introduce the fluctuating internal local magnetic induction $\mathbf{B}_{int} = \overline{\mathbf{B}}_{int} + \Delta \mathbf{B}$ which acts on each particular dislocation – obstacle bond. It means that we have to average equation (11) over various values and orientations of the vector $\Delta \mathbf{B}$. This averaging for the case of an easy axis ferromagnet is carried out in Appendix), so one gets

$$\frac{\sigma_c(T)}{\sigma_{c0}} = \overline{f}(\overline{B}, \langle \Delta B^2 \rangle) \left[1 - \left(\frac{T}{T_0} \right)^{\frac{2}{3}} \right]^{\frac{3}{2}}. \tag{12}$$

The critical stress appears now to be a function of the average spontaneous magnetization and of its fluctuations. The temperature dependence of both these quantities is determined by equations (6), (7), and (9). As a result we now have an equation for the temperature dependence of the critical stress in the vicinity of the Curie temperature.

Figure 1 presents a comparison of the temperature dependence of the critical stress in Ni calculated by means of equation (12) with the available experimental data. The theoretical curve is plotted using the following parameters for Ni: $T_C = 627$K, $M_0 = 510$G, $p = 0.606$ (Kittel, 1986), $\beta = 0.33$, $\gamma = 1.33$ (Kadanoff etal, 1967). The effective number, p, of Bohr
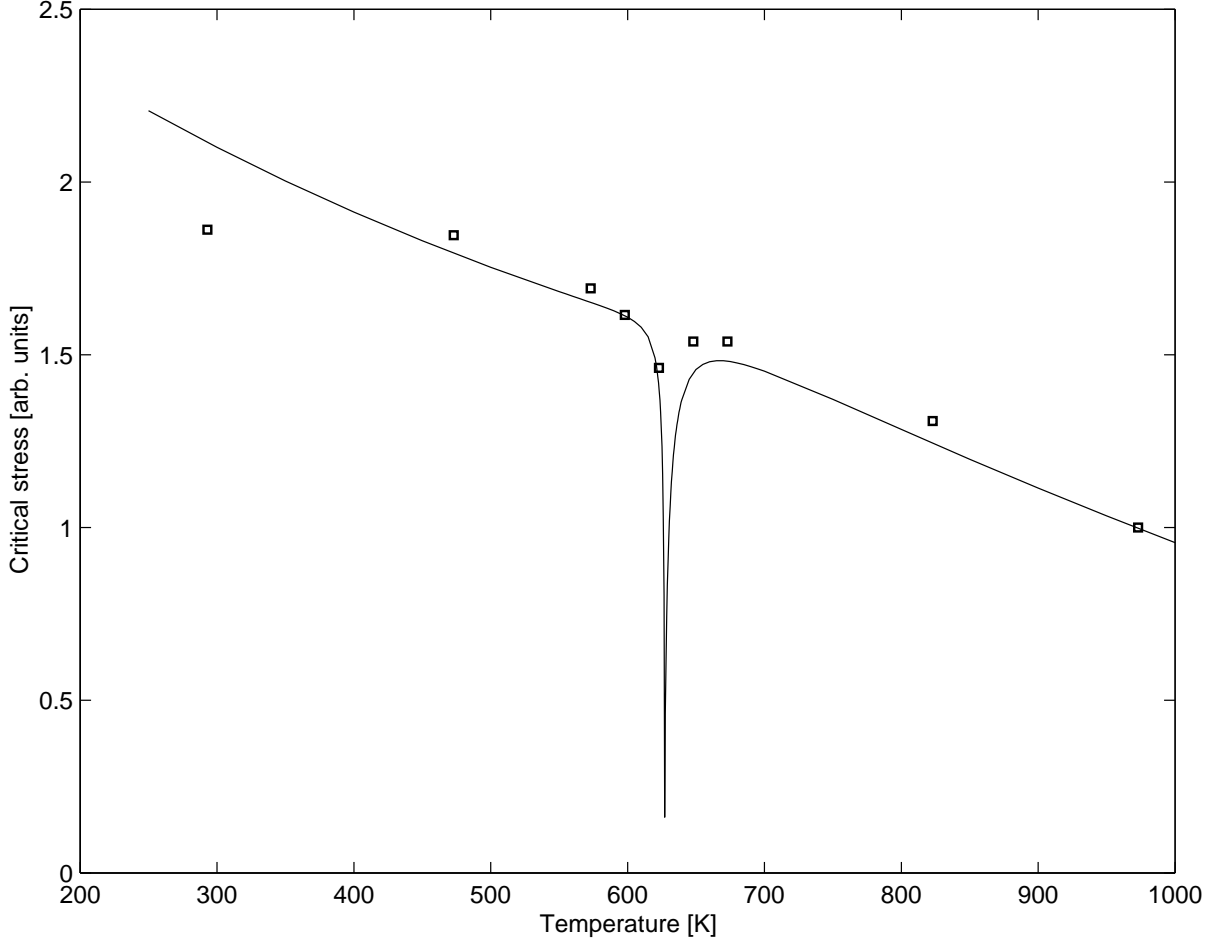
FIG. 1: Temperature dependence of the critical stress in Ni.

magnetons in Ni is rather small and, hence, one gets a small Curie constant $C = 7 \times 10^{-3} K$. The remaining parameters, $T_0 = 2070$K, $B_0 = 0.49$T, $V^* = 2.64 \times 10^{-22}$cm^3, are determined by fitting equation (12) to the experimental data by Zackay and Hazlett (1953) for their samples with 2% deformation. One observes a reasonable general agreement between the theory and experiment. However, the measurements were carried out with a ~ 20K interval between the points. As a result the sharp minimum in the temperature dependence of the critical stress near the Curie point, predicted by our theory, might have been overlooked. A relatively small, ~ 15%, decrease of the critical stress in this region is indicative of a possible existence of a sharper minimum in this temperature range.

IV. TEMPERATURE DEPENDENCE OF THE MICROHARDNESS: GADOLINIUM AS A TEST CASE

This section discusses the behavior of crystal microhardness near the Curie point. The results are compared with the available experimental data on gadolinium. Currently there is no microscopic theory of microhardness, and hence, no microscopic theory of the influence of a magnetic field on it. Nevertheless, the dependence of the microhardness on a magnetic field can be estimated by means of the following simple considerations. It is known that the microhardness H varies inversely proportionally to the plasticity — the higher the plasticity, the lower the hardness. On the other hand, the plasticity is directly connected to the dislocation path length. The magnetic field dependence of $L(B)$ is given by equation (1). Hence, we assume that the average microhardness of a crystal as

$$H = H_0 \frac{L_0}{L(B)} \tag{13}$$

where H_0 is the microhardness in the absence of a magnetic field.

Assuming an isotropic distribution of the orientations one gets

$$H = H_0 \overline{f}(\overline{B}, \langle \Delta B^2 \rangle) \tag{14}$$

where the function $\overline{f}(\overline{B}, \langle \Delta B^2 \rangle)$ is calculated in Appendix. Similarly to the previous section, we use equations (6), (7), and (9) in order to get equation (14) for the temperature dependence of the microhardness in the vicinity of the Curie temperature.

A comparison of this theoretical result with the experimental data available for the microhardness of gadolinium is presented in Figure 2. The Curie constant C for gadolinium can be found using equation (8) and the values of the relevant parameters: $n = 3.02 \times 10^{22} \text{cm}^{-3}$, $p = 7.10$, $M_0 = 2010\text{G}$ (Kittel, 1986), which lead to $C = 0.33\text{K}$. The Curie temperature for gadolinium is $T_C = 292\text{K}$ (Nabutovskaya, 1969). The critical indices are $\beta = 0.3265$ and $\gamma = 1.239$ (Aliev etal, 1988).

The value of the parameter B_0 could have been possible to determine from the magnetic field dependence of the plasticity. Unfortunately, we do not know about such measurements in gadolinium. However, magneto- (Al'shits etal, 1990) and electroplastic (Okazaki etal, 1979) effects have been measured in zinc and titanium, who, similarly to gadolinium, possess hcp structures. Fitting these results to the theory (Molotskii, 2000) provides $B_0 = 0.70\text{T}$ for Zn and $B_0 = 0.94\text{T}$ for Ti.
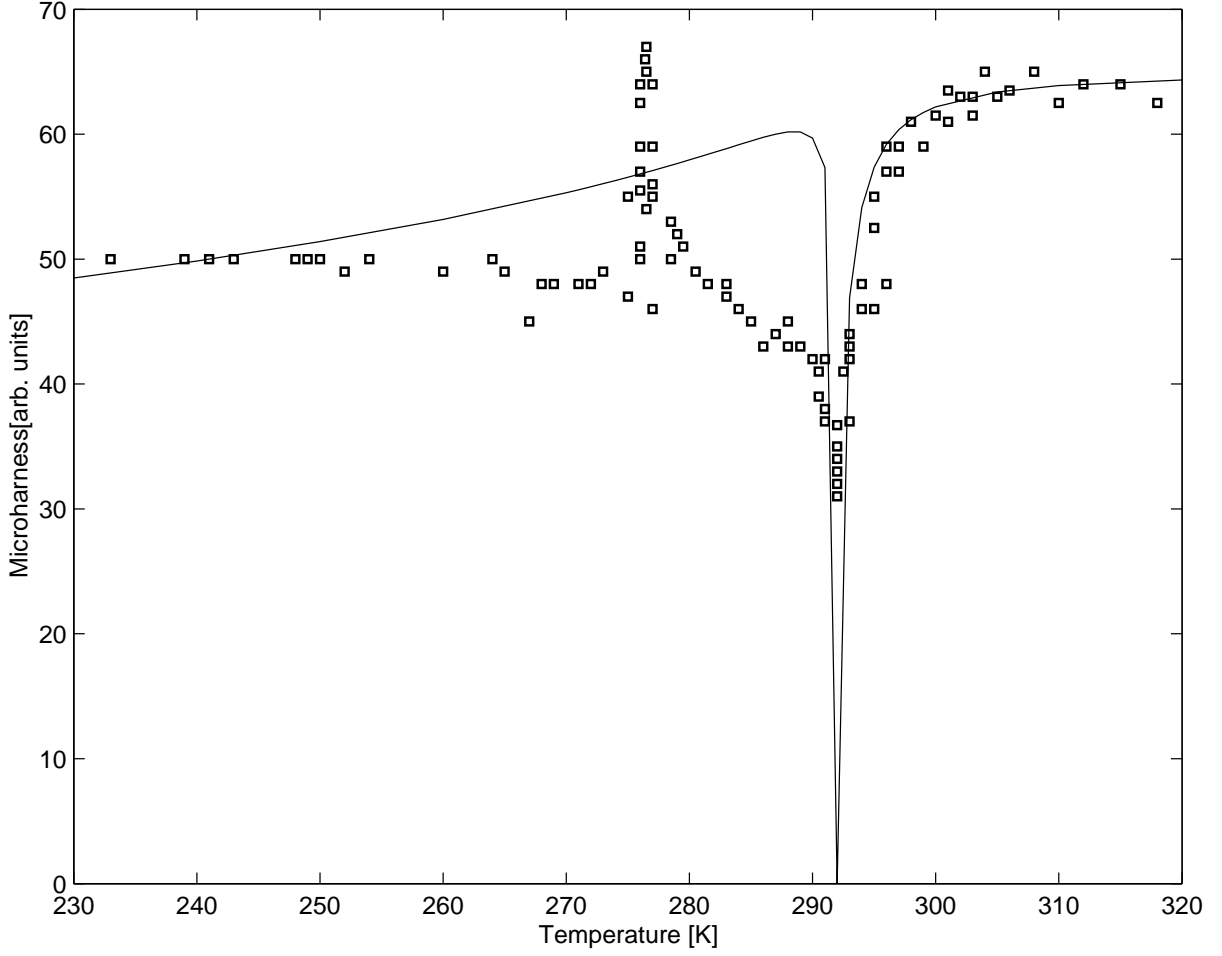
FIG. 2: Temperature dependence of the microhardness of Gd.

The theoretical curve in Figure 2 uses the values B_0 and V^* as fitting parameters. The obtained parameter $B_0 = 0.87$T for gadolinium appears to be closed to its experimentally measured values for Zn and Ti. The characteristic volume is chosen $V_0 = 1.85 \times 10^{-20}$cm^3. It corresponds to a sphere with the radius 17Å, about four interatomic spacings. The results are not very sensitive to the variation of this volume, which can be chosen smaller (up to 2×10^{-21}cm^3) without damaging the overall agreement.

V. CONCLUSION

Accounting for the internal magnetic induction of ferromagnets and for its fluctuations leads us to the conclusion that various plastic properties of crystals should be singular near the Curie point. The critical behavior of the magnetic susceptibility is transferred to the

temperature dependence of the critical stress and microhardness. It would be interesting to measure other plasticity characteristics, such as dislocation path length L or plastic strain rate $\dot{\varepsilon}$ near the Curie point in order to look for a possible singular behavior, which follows from the theory proposed in this paper. For example, one may expect that carrying out measurements similar to those of Zackay and Hazlett, 1953, on the temperature dependence of the critical stress in Ni, but with a smaller temperature interval a sharp minimum at $T = 627$K may be observed.

An external magnetic field is known to smear out the phase transition, to decrease the magnetic susceptibility, and to suppress fluctuations near T_C. As shown by Dan'kov etal (1998) a 0.5T magnetic field diminishes the magnetic susceptibility near the Curie point in gadolinium by an order of magnitude. This should lead to a nearly complete suppression of the magnetization fluctuations and, hence, to a suppression of the singular temperature dependence of microhardness. As for nickel, a stronger field about 3T (Hischler and Rocker, 1966) is necessary to suppress the fluctuations, hence correspondingly higher field will be necessary to influence the singular behavior of the critical stress in nickel near the Curie point.

We may conclude that an experimental observation of a suppression of the above singularities in the temperature dependencies of plasticity characteristics in a magnetic field will be an important experimental evidence in favour of the theoretical approach to the plasticity of ferromagnets presented in this paper.

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APPENDIX A: AVERAGING FOR AN EASY AXIS CUBIC CRYSTAL

The averaging is carried out assuming a cubic symmetry. It is certainly relevant to Ni. As for the hexagonal Gd, its anisotropy is rather weak and the result obtained here, will be also applied for this crystal. We have to average the quantity

$$f(B) = \frac{1}{1 + \dfrac{\mathbf{B}^2}{B_0^2}}, \tag{A1}$$

where $\mathbf{B} = \overline{\mathbf{B}} + \Delta\mathbf{B}$, over the distribution (4). Disregarding a possible anisotropy of the fluctuations of the magnetic induction in the vicinity of the dislocation – obstacle bonds this

averaging can be presented as

$$\langle f(B) \rangle \equiv \overline{f}(\overline{B}, \langle \Delta B^2 \rangle) = \frac{1}{(2\pi)^{1/2} V^* \langle \Delta B^2 \rangle^{3/2}} \int_{V^*}^{\infty} dV \int_0^{\infty} \Delta B^2 d\Delta B \int_{-1}^1 \frac{d\cos\theta}{2} \times$$

$$\frac{1}{1 + \frac{1}{B_0^2}(\overline{B}^2 + \Delta B^2 + 2\overline{B}\Delta B \cos\theta)} \exp\left\{ -\frac{\Delta B^2 V}{2\langle \Delta B^2 \rangle V^*} \right\} \tag{A2}$$

Now integration over ΔB is carried out by parts and the averaging (A2) is represented as

$$\overline{f}(\overline{B}, \langle \Delta B^2 \rangle) =$$

$$\frac{1}{2(2\pi)^{1/2} \langle \Delta B^2 \rangle^{1/2}} \int_{V^*}^{\infty} dV \frac{1}{V} \int_{-\infty}^{\infty} d\Delta B \frac{1}{1 + \frac{1}{B_0^2}(\overline{B} + \Delta B)^2} \exp\left\{ -\frac{\Delta B^2 V}{2\langle \Delta B^2 \rangle V^*} \right\} +$$

$$\frac{1}{2(2\pi)^{1/2} \langle \Delta B^2 \rangle^{1/2} \overline{B}} \int_{V^*}^{\infty} \frac{dV}{V} \int_{-\infty}^{\infty} d\Delta B \frac{\Delta B}{1 + \frac{1}{B_0^2}(\overline{B} + \Delta B)^2} \exp\left\{ -\frac{\Delta B^2 V}{2\langle \Delta B^2 \rangle V^*} \right\} \tag{A3}$$

or

$$\overline{f}(\overline{B}, \langle \Delta B^2 \rangle) = \frac{1}{2\sqrt{2\pi}} \int_1^{\infty} \frac{dv}{v} \int_{-\infty}^{\infty} dx \frac{a^2}{a^2 + (b+x)^2} \left(1 + \frac{x}{b}\right) \exp\left\{ -\frac{x^2 v}{2} \right\} \tag{A4}$$

where the notations $a = B_0/\sqrt{\langle \Delta B^2 \rangle}$, $b = \overline{B}/\sqrt{\langle \Delta B^2 \rangle}$, $v = V/V^*$, and $x = \Delta B/\sqrt{\langle \Delta B^2 \rangle}$ are introduced. The identity

$$\frac{1}{a \pm i(b+x)} = \int_0^{\infty} dp \exp\{-p[a \pm i(b+x)]\} \tag{A5}$$

is substituted into (A4). Now integration over x is carried out and a new integration variable $z = p/\sqrt{v}$ is introduced instead of v. After that, p is substituted for pa and one gets

$$\overline{f}(\overline{B}, \langle \Delta B^2 \rangle) = \frac{B_0}{\sqrt{\langle \Delta B^2 \rangle}} \int_0^{\infty} dp \times$$

$$\left\{ \sqrt{\frac{\pi}{2p}} e^{-p} \Phi\left(\frac{\sqrt{\langle \Delta B^2 \rangle} p}{B_0 \sqrt{2}}\right) \left[\cos\left(\frac{\overline{B}p}{B_0}\right) - \frac{B_0}{4\overline{B}p} \sin\left(\frac{\overline{B}p}{B_0}\right)\right] + \frac{B_0}{4\overline{B}p} e^{-p - \frac{\langle \Delta B^2 \rangle p^2}{2B_0^2}} \sin\left(\frac{\overline{B}p}{B_0}\right) \right\} \tag{A6}$$

where

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt \tag{A7}$$

is the probability integral.

The value of $\overline{f}(\overline{B}, \langle \Delta B^2 \rangle)$ can be analytically estimated in the limit of small fluctuations, when $\sqrt{\langle \Delta B^2 \rangle} \ll B_0$. The integral (A6) converges for small values of p and the expansion $\Phi(x) \approx 2x/\sqrt{\pi}$ can be used. It converges to a rather obvious result

$$\overline{f}(\overline{B}, \langle \Delta B^2 \rangle) = \frac{B_0^2}{B_0^2 + \overline{B}^2}. \tag{A8}$$

For large fluctuations when $\sqrt{\langle \Delta B^2 \rangle} \gg B_0$ the integration (A6) includes a broad range of large values of p. The probability integral $\Phi(x)$ rapidly converges to one for $x \gg 1$. Then we may exclude the range $p < p_0 = \dfrac{B_0 \sqrt{2}}{\sqrt{\langle \Delta B^2 \rangle}}$ whose contribution to the integral is of the order of $\left(\dfrac{B_0}{\sqrt{\langle \Delta B^2 \rangle}} \right)^2$ and integrate for $p > p_0$. Then one gets

$$\overline{f}(\overline{B}, \langle \Delta B^2 \rangle) = -\frac{B_0}{2\sqrt{\langle \Delta B^2 \rangle}} \sqrt{\frac{\pi}{2}} \left[\mathrm{Ei}(-\sqrt{2}\frac{B_0 + i\overline{B}}{\sqrt{\langle \Delta B^2 \rangle}}) + \mathrm{Ei}(-\sqrt{2}\frac{B_0 - i\overline{B}}{\sqrt{\langle \Delta B^2 \rangle}}) \right] \approx$$

$$\sqrt{\frac{\pi}{2}} \frac{B_0}{\sqrt{\langle \Delta B^2 \rangle}} \left[\ln(\frac{\langle \Delta B^2 \rangle}{B_0^2 + \overline{B}^2}) - C - \frac{1}{2} \ln 2 \right] \tag{A9}$$

where $C = 0.577$ is the Euler number. $\mathrm{Ei}(x)$ is the integral exponential function.

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